

Magnetic Refrigeration for Maser Amplifier Cooling

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The development of a multifrequency upconverter-maser system for the DSN has created the need to develop a closed-cycle refrigerator (CCR) capable of providing more than 3 watts of refrigeration capacity at 4.5 K. In addition, operating concerns such as the high cost of electrical power consumption and the loss of maser operation due to CCR failures require that improvements be made to increase the efficiency and reliability of the CCR. One refrigeration method under consideration is the replacement of the Joule-Thomson expansion circuit with a magnetic refrigerator. Magnetic refrigerators can provide potentially reliable and highly efficient refrigeration at a variety of temperature ranges and cooling powers. This paper summarizes the concept of magnetic refrigeration and provides a literature review of existing magnetic refrigerator designs which have been built and tested and that may also be considered as possibilities as a 4 K to 15 K magnetic refrigeration stage for the DSN closed-cycle refrigerator.

I. Introduction

Efficient long-life refrigeration techniques are being developed for the DSN masers to improve the present closed-cycle refrigerator technology. One area of investigation is to find an alternative to the Joule-Thomson refrigeration process in the DSN closed cycle refrigerators (CCRs). In the CCR, as in virtually all mechanical refrigerators which are capable of the liquefaction of helium, a Joule-Thomson (J-T) circuit is added to an expansion engine to serve as the final stage of cooling to produce the desired refrigeration at the liquid helium (LHe) temperatures. The J-T circuit, while having no moving parts or seals, depends critically on extreme high gas purity to obtain long-term continuous operation. In fact, a major reason for present CCR failures is due to gas impurities which plug the J-T circuit. Because the J-T circuit and the precooled expansion engine circuit are essentially independent

of each other, it is important to determine whether other refrigeration cycles can be used in place of the J-T circuit which will match or surpass the J-T circuit in terms of efficiency, reliability, temperature span achievable, and ease of construction and operation. One such refrigeration mode under investigation is the cooling by adiabatic demagnetization.

Adiabatic demagnetization of a paramagnetic salt (or magnetic refrigeration) was chosen as the possible replacement for the J-T circuit due to the following potential advantages over the J-T circuit.

- (1) High efficiencies are attainable (more than 70% of Carnot efficiency in comparison to 20-25% for the J-T expansion stage as measured for a DSN 1-watt CCR (Ref. 1)).

- (2) The refrigeration capacity per unit volume of working material is large as high density solids rather than the low density gases are used.
- (3) Potential high reliability can be expected since only a few slow moving parts are required.
- (4) The paramagnetic material is relatively inexpensive (e.g., gadolinium gallium garnet costs about \$225/lb).

There are several disadvantages which must also be considered.

- (1) The need for a high field superconducting magnet that must be maintained at liquid helium temperatures.
- (2) Stray magnetic fields which will require shielding to protect the maser.
- (3) The use of low temperature seals for moving parts.

The development work on cyclic magnetic refrigeration to date has shown great promise for its application as a viable means of providing efficient and reliable refrigeration. Their proposed uses include:

- (1) Laboratory material studies in the μ kelvin range through adiabatic demagnetization of the salt pill's nuclear spins (Ref. 2.).
- (2) Spacecraft operations to cool detectors and instruments (Refs. 3, 4).
- (3) Large-scale refrigeration for superconductivity application in power generation and transmission (Ref. 5).
- (4) Complete magnetic refrigeration systems operating between 300 K and 20 K (or 4 K) for the production of liquid H_2 (or liquid He) (Ref. 6).

It is interesting to note that none of the material refrigerators developed to date have been used to produce 4 K refrigeration. The major development work thus far has centered around room temperature devices where the use of superconducting magnets could be avoided, or below 4 K where a liquid helium bath could be used to submerge the superconducting magnet and also be used for the high-temperature heat reservoir.

Van Geuns (Ref. 7) first proposed the idea of a 4-15 K magnetic refrigeration stage as early as 1966 to find a more efficient alternative to the J-T circuit of a closed-cycle refrigerator. Although the results of the analysis of the proposed refrigerator indicated that the prospects for the magnetic refrigerator were promising, no follow-up experimental work has been reported in the literature to our knowledge. Since that time there have been several studies of paramagnetic materials for use in a 2-20 K temperature range (Refs. 8, 9),

and there have been more proposed designs for a 4-15 K magnetic refrigerator (Refs. 10-12), but to date no experimental devices have been developed.

In the next section, the concept of magnetic refrigeration will be described and possible thermodynamic cycles available for a magnetic refrigeration stage will be introduced. A brief discussion will be made of several magnetic refrigerators that have been reported on in the literature and how they may be used as a 4-20 K refrigerator stage for use in a DSN CCR. We will also introduce some of the design constraints that must be considered if a magnetic refrigeration stage is to be used.

II. Principles of Magnetic Refrigeration

Magnetic cooling is a consequence of the fact that at a fixed temperature the entropy of a system of magnetic dipoles is lowered by the application of a magnetic field. In the absence of an external magnetic field the magnetic moments of the electron spin system are randomly oriented, with all orientations equally probable. When a magnetic field is applied, the dipole moments tend to align parallel to the field, thereby increasing order in the system, or equivalently, decreasing its entropy. In doing so the moments go into lower energy levels, giving off excess energy to the crystal lattice of the paramagnetic material in the form of a heat of magnetization. This heat energy is removed from the material by holding it in contact to a constant-temperature reservoir. Now by thermally isolating the material and removing the field isentropically, the dipoles again become randomly oriented. But in doing so, energy must be drawn out of the material's lattice, thereby lowering the temperature of the material.

The entropy-temperature diagram (or S, T-diagram) of a paramagnetic material is shown in Fig. 1. The entropy curves at several constant magnetic fields have been shown. The temperature T_0 is defined as the temperature at which the zero magnetic field entropy per mole of material is equal to $R \ln (2J + 1)$ (see Fig. 1). At temperatures above T_0 , the T^3 contribution to the entropy by the lattice vibrations becomes appreciable. Below T_0 the molar entropy depends little on the lattice entropy and thus remains fairly temperature-independent until the interaction energy E of the ions becomes an appreciable fraction of the average thermal energy kT . Spontaneous ordering of the dipoles then occurs and this lowers the entropy. This can be characterized by a magnetic ordering temperature $\Phi \cong E/k$. Here the specific heat of the material also shows a pronounced maximum (since it varies as $T \cdot \partial S / \partial T$). Between Φ and T_0 the entropy of the spin system depends most strongly on the magnetic field, and in this region the adiabatic demagnetization method is most effective.

III. Thermodynamic Cycles

The concept of cyclic magnetic refrigeration can best be described with the aid of Fig. 2. In phase 1 of the cycle, thermal switch S2 is open and thermal switch S1 is closed so that the working material M is in good thermal contact with the heat reservoir R during the application of a magnetic field. This permits the heat of magnetization $Q = T\Delta S$ to be removed from the material while the temperature of the material T_M remains nearly constant at the reservoir temperature T_R . In phase 2, both thermal switches are opened to thermally isolate the working material and the field is reduced until T_M is lowered to the heat source temperature T_S . In phase 3 switch S2 is closed to permit thermal contact between the working material and the heat sources while the field is reduced to zero. Heat energy is extracted from the source while the final demagnetization is lowering T_M . This process establishes a somewhat lower source temperature T_S . Phase 4 represents the last step in the magnetic cycle. During this phase, S1 and S2 are opened to isolate the material and the field is increased until T_M is increased to T_R . The magnetic cycle can be repeated by returning to phase 1 of the cycle and again applying the full field to the working material. The temperature T_S will continue to fall with each cycle of the refrigerator until the net refrigeration of the working material is balanced out by the heat leak to the source.

The magnetic refrigerator in the above example operated on the basis of the Carnot cycle (two isothermal steps and two adiabatic steps). It is an easy cycle to execute but has a limited temperature span while requiring a large field change. Other magnetic analogs to the gas thermodynamic cycles exist which are more difficult to carry out, but they are capable of larger temperature spans while requiring smaller magnetic field changes. They are: the Stirling cycle (Ref. 6), which consists of two isothermal and two isomagnetization steps; the Ericsson cycle (Ref. 6), which consists of two isothermal and two isofield steps; and the Brayton cycle (Ref. 6), which consists of two adiabatic and two isofield steps. The four cycles are shown in Fig. 3, but they do not represent the only refrigeration cycles possible. The operation of a magnetic refrigerator based on one of these thermodynamic cycles will depend largely on the design of the refrigerator, the ease of implementation and the environment in which the refrigerator will work.

IV. Approaches to Actual Magnetic Refrigeration Devices

A. Two-Switch Devices

It was the Nobel prize winning work of Giauque and MacDougall (Ref. 13) in 1933 which demonstrated the idea

of magnetic refrigeration to cool from 3.5 to 0.5 K. Subsequent magnetic refrigeration work was able to produce temperatures to below 1 mK. Until recently, the method of adiabatic demagnetization was limited to so-called "one-shot operations" where the paramagnetic material was isothermally magnetized and followed by an adiabatic demagnetization to obtain temperatures below 1 mK for limited periods of time.

In 1954 Heer et al. (Ref. 14) developed the first magnetic refrigerator to operate in a continuous mode to provide a fairly constant low temperature. The device used superconducting thermal valves (as diagrammed in Fig. 1) and a high heat capacity material for the low-temperature reservoir to smooth out the low-temperature fluctuations. This refrigerator was capable of producing $7 \mu\text{W}$ of refrigeration at 0.26 K. Arthur D. Little, Inc. commercially marketed a few magnetic refrigerators modeled after this design (Ref. 15) until ^3He became commercially available in sufficient quantities to use as a refrigerant in this temperature range.

The next refinement was by Zimmerman et al. (Ref. 16) in 1962, by using a superconducting solenoid. This refrigerator was capable of producing $100 \mu\text{W}$ of refrigeration at 0.26 K. More recently Rosenblum et al. (Ref. 17) used a similar device to produce $0.1 \mu\text{W}$ of refrigeration at the much lower temperature of 10 mK. All of these refrigerators were low power devices and limited to temperatures below 1 K because of the superconducting switches used. Steyert (Ref. 12) recently proposed a 2-10 K refrigerator which uses magnetoresistive switches (Ref. 18) with large switching ratios to produce several watts of refrigeration at 2 K using a Carnot cycle. No such refrigerator has been built to date.

B. Reciprocating Devices

The reciprocating magnetic refrigerator requires the complex movement of two of the following components: the magnet, the magnetic material, or the regenerator fluid. Described simply, the working material passes through the regenerative column of fluid in which a temperature gradient has been established after numerous cycles. The regenerative fluid does the work of passing the heat from the heat source to the heat sink. The mixing of the fluid across the temperature gradient by the movement of the material is the key area of concern for the design of the device. An illustration of one such refrigerator (Ref. 19) is shown in Fig. 4.

Three reciprocating magnetic refrigerators have been built and tested since Van Geuns (Ref. 7) first proposed the idea of a 4-15 K reciprocating magnetic refrigeration stage to replace the J-T circuit of his closed cycle refrigerator. Brown (Ref. 19) utilized a magnetic Stirling cycle to demonstrate a 47 K temperature span between 272 and 319 K. He used gadolinium

metal as the working material which passed through a regenerative column of water and ethyl alcohol. Barclay et al (Ref. 20) used a salt pill of compressed $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$ to pump heat from 2 to 4 K. Using a magnetic Ericsson cycle at a 1/60 Hz rate, they were able to develop 52 mW of refrigeration capacity at 2 K in their initial tests. Delpuech et al. (Ref. 21) built a 2 to 4 K double-acting reciprocating device in which two identical $\text{Gd}_2(\text{SO}_4)_3$ magnetic elements on a reciprocating drive shaft alternately entered and left the magnetic field. Incorporating this arrangement with a magnetic Ericsson cycle, they were able to attain 570 mW of refrigeration capacity at 2.1 K at a 0.3-Hz cycle rate. In later experiments using $\text{Gd}_2\text{Ga}_5\text{O}_{12}$ magnetic elements, they were able to achieve 1.2 W at 1.8 K with a 0.95 Hz rate (Ref. 22).

C. Rotating Wheel Devices

The wheel-concept magnetic refrigeration device uses the magnetic material to form the rim of the wheel. It has a simple axial drive but requires a more complicated magnetic field profile since one portion of the wheel must be in a large field while the opposite side must be in a low magnetic field. However, because one portion of the rim is constantly rotating into or out of the magnetic field, the wheel design does provide continuous refrigeration. Figure 5 illustrates the concepts of both a Carnot-cycle and a Stirling-cycle wheel design.

Steyert and coworkers (Refs. 5, 24) have developed several rotating wheel designs which have operated at either room temperature or between 2 and 4 K. The prototype low-temperature Carnot-cycle wheel design (Ref. 24) used $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$ as the working material and was capable of pumping 0.5 W of heat from 2.75 K to the 4.2 K heat sink. The wheel rotated at 0.77 Hz. The capacity and source temperature varied with the rotation rate of the wheel.

As a feasibility study for the use of low-temperature magnetic refrigeration technology to provide refrigeration for superconducting applications in power generation and transmission, Barclay and Steyert (Ref. 5) built a room-temperature wheel with an objective of understanding the device in an experimentally convenient temperature range. Their prototype gadolinium metal wheel operated through a 1.2 tesla field change and provided 0.5 kW of continuous refrigeration over a 7 K temperature span while rotating at a 0.5 Hz rate. This Stirling-cycle wheel device used water as the regenerative fluid.

D. Rotating Anisotropic Crystal

Barclay (Ref. 11) proposed this magnetic refrigerator design based on the anisotropic nature of certain single-crystal paramagnetic materials. The anisotropic material, such as DyPO_4 , is rotated in a constant magnetic field causing a

drastic change in the material's magnetic entropy. The temperature changes associated with the changes in the paramagnetic ion polarization are used directly to heat or cool a heat exchange fluid. Each 360° rotation of the crystal represents a double execution of the thermodynamic cycle. Barclay describes a Carnot-cycle 4 to 20 K rotational refrigeration device that is capable of producing 1 W of refrigeration. A conceptual drawing of the device is shown in Fig. 6.

E. Active Magnetic Regenerator Device

The active magnetic regenerator is a device composed of layers of one or more magnetic materials having different Curie temperatures (depending on the temperature span desired) that are thermodynamically cycled like the ordinary lead regenerator of the CCR, except in this case the temperature of the materials can be changed by the application or removal of a magnetic field. Each of the different materials executes a small Brayton cycle near its Curie temperature. Depending on the number and types of different materials used, a temperature span of 10-220 K may be achieved. This concept was proposed by Barclay and Steyert (Ref. 10) and is still in the development stage. A prototype room-temperature device has been built and tested (Ref. 5) in which initial results show an 18 K temperature span using only gadolinium metal as the magnetic material.

V. A Magnetic Refrigeration System

The present and future requirements of the CCR system for the DSN are listed in Table 1. With the development of a multifrequency upconverter maser system there exists the need to develop a new CCR capable of providing 3-4 watts of refrigeration power at 4.5 K. A scaled-up version of the present DSN 1W CCR, capable of more than 3 W of refrigeration power at 4.5 K, is presently being tested. Also under investigation is the development of a 4-15 K magnetic refrigeration stage which will replace the J-T expansion circuit to provide the final stage of cooling in the CCR.

The reasons for considering a 4 to 15 K magnetic refrigeration stage are the advantages it has over the J-T circuit. This replacement eliminates the CCR failures that are a direct result of J-T circuit blockage due to gas impurities. The refrigeration power per unit volume of paramagnetic material is large (since we are dealing with a solid as the working substance rather than a gas, and the density of the solid is more than an order of magnitude greater than that of the compressed gas). This should facilitate the use of a small and compact design. The magnetic cycle itself can operate at nearly 100% efficiency; when the losses associated with the magnetic stage are included, the Carnot efficiency of the magnetic stage should still be on the order of 70% or more.

The magnetic refrigerator stage will require some moving parts, but as they will be moving at slow speeds the overall CCR reliability should not be impaired.

In order to incorporate a 4-15 K magnetic refrigeration stage into a closed-cycle refrigerator, the refrigerator components and their constraints must first be identified and fully understood. The major system components of the magnetic refrigerator are listed in Table 2. While the exact magnetic thermodynamic cycle to be used will play a part as to what the final design configuration of each component will be, it is nevertheless important to identify the main components and describe their general details. In what follows, we will discuss the pertinent points of the first four items listed in Table 2.

A. Magnetic Working Material

It is necessary to select the working material and the magnetic cycle which will best operate over the temperature span desired. Through the temperature span of interest, both the entropy and the heat capacity of the spin system of the material must be large compared to those of its lattice system so that the low spin temperature attained during a demagnetization is not appreciably curtailed by the internal heat load of the lattice. The magnetic ordering temperature of the material should be a degree or two below the lowest refrigeration temperatures desired because the magnetic entropy changes with external field become very small below this point. Other important characteristics to be considered must be its thermal, chemical and physical properties. Finally, cost, availability and machinability are also important considerations.

A study of paramagnetic materials suitable for magnetic refrigeration in the 2-20 K range has been conducted by Barclay and Steyert (Ref. 9) for JPL as part of the overall R&D program on magnetic refrigeration at the Los Alamos Scientific Laboratory. The study was restricted to gadolinium compounds. In particular gadolinium gallium garnet ($\text{Gd}_2\text{Ga}_5\text{O}_{12}$ or GGG) was shown to be an excellent choice as the working material in the 4-15 K range because of its high thermal conductivity, its very low lattice heat capacity and its low ordering temperature (~ 0.8 K). The material is the standard substrate material used in magnetic bubble domain technology and therefore is readily available. Grown as a single crystal boule it is also very machinable. This material is presently being examined at JPL and results will be forthcoming.

B. Heat Exchange Mechanism

During the magnetic cycle, heat is periodically exchanged between the working material and the heat sink or heat

source. There are two distinct ways in which this heat exchange can take place: through a thermal switch such as is needed in the isothermal stage of a Carnot cycle or through a regenerative fluid flow as would be used in the Stirling cycle, for example. Thermal switches require large switching ratios if they are to work efficiently. Two likely candidates would be magnetoresistive switches (Ref. 18), such as single crystals of beryllium, with switching ratios of 1000 at temperatures as high as 15 K, or helium gas gap switches which are actuated quickly by electrically heating charcoal to change the gas pressures. Of main importance is that no regenerative fluid is used since the field is changed by varying the magnet. However, this may cause serious problems for a nearby maser unless adequate shielding is provided. As another heat exchange mechanism, a heat exchange fluid such as high pressure helium gas can also be used to couple the working material to the hot or cold sources and to effect the regeneration. Fluid pumps will be required to operate at low temperatures to force the circulation of the fluid. It is perhaps better still to use the working material as the regenerator as in the active magnetic regenerator device. The magnetic material could be shaped in small spheres and have the helium forced through the regenerator bed to attain the necessary heat transfer.

C. Magnetic Material Drive Mechanism

The work for the magnetic refrigeration stage is provided by the drive motor. For a reciprocating design a force compensation mechanism must be utilized to pull the material in and out of the field because of the large forces involved. A more complicated cam and drive shaft are required if both the magnet and the magnetic material are to move. For both the rotating wheel and the rotating anisotropic crystal a simple axial motion of the drive shaft is used. The wheel provides its own force compensation by its continuous geometry; therefore the force from the drive shaft's torque will be smaller than the force required for the reciprocating drive units.

D. Superconducting Magnet

The biggest disadvantage in a magnetic refrigeration stage for the CCR is the need for a high field superconducting magnet. Stray fields from this magnet would pose a serious problem for maser operation unless effective shielding was provided. The shielding should be adequately accomplished through the use of high permeability or superconducting materials.

Another problem to be overcome is how to cool the magnet initially. The magnet can be cooled by an initial LHe

transfer but this method is not very feasible in light of the CCR being installed on the antennas. The magnet will most likely be precooled to 12-15 K by the two-stage expansion engine as the J-T circuit is now. From there, two options are possible: to use a Simon liquefier (expansion of stored high pressure helium gas that has been precooled to under 15 K) to produce sufficient LHe to cool the magnet; or to start the magnetic cycle at low fields at 12-15 K and have the magnetic stage cool itself down. The latter choice requires a high T_c superconducting wire such as Nb_3Sn . In either situation, once the magnetic refrigerator is operating, the small parasitic heat load required by the magnet operating in the persistent mode will require only a small fraction of the total large refrigeration capacity of the magnetic refrigeration stage.

VI. Conclusions

The idea of using magnetic refrigeration to provide the 4 K refrigeration for the DSN maser has been introduced. The main feature of the magnetic refrigerator is its potential high efficiency (more than 70% of Carnot efficiency can be expected). Additionally, because it uses solid materials it is inherently orientation-independent. This, and other design constraints laid on the refrigerator by the operating maser system on the antenna have been listed. Several different refrigerator devices have been presented as possible designs for a 4-15 K magnetic refrigeration stage. The question to be answered will be which type of magnetic refrigeration device using which thermodynamic cycles will prove to be optimal for our present and future refrigeration requirements.

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Table 1. CCR System requirements for maser cooling

Reliable
Efficient
Multiyear lifetime
Unattended operation
Rapid cooldowns
1-4 watt cooling capacity
Compact
Magnetic field isolation of maser package
Low microphonics
mK temperature stability
Orientation independence
Continued operation during power failures

Table 2. Major components of a magnetic refrigerator system

Solid magnetic working material
Heat exchange mechanism
Magnetic material drive mechanism
Superconducting magnet
Dewar
15 K refrigerator precooler

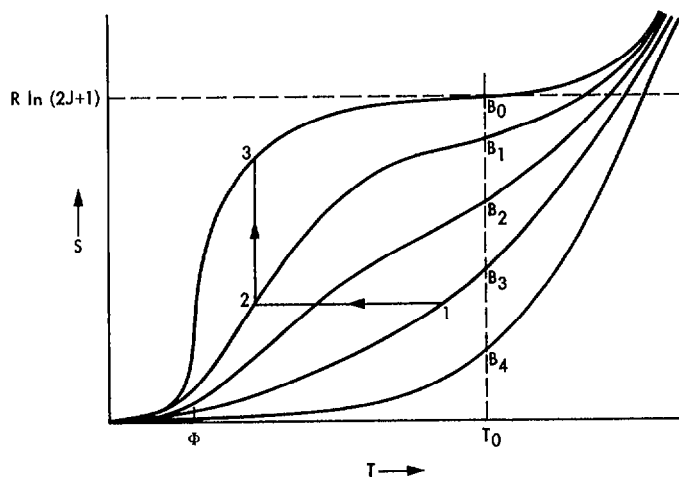


Fig. 1. S, T-diagram for a typical paramagnetic material showing the entropy vs temperature curves for several applied magnetic fields. Process 1 \rightarrow 2 shows an isentropic (adiabatic) demagnetization which decreases the temperature of the material. Process 2 \rightarrow 3 shows an isothermal demagnetization whereby the material absorbs heat from low-temperature heat source. Temperatures Φ and T_0 are defined in the text

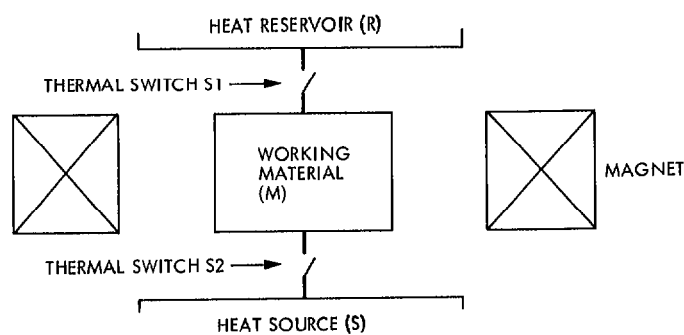


Fig. 2. Schematic diagram of a conventional magnetic refrigerator. The cyclic operation of this refrigerator is discussed in the text

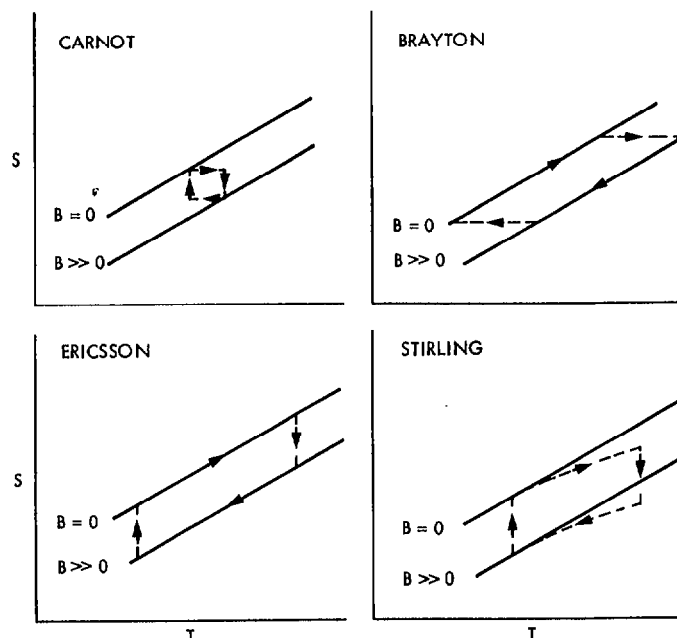


Fig. 3. Four magnetic cycles that can be used to make a cyclic magnetic refrigerator. The processes of each cycle are mentioned in the text (taken from Ref. 6)

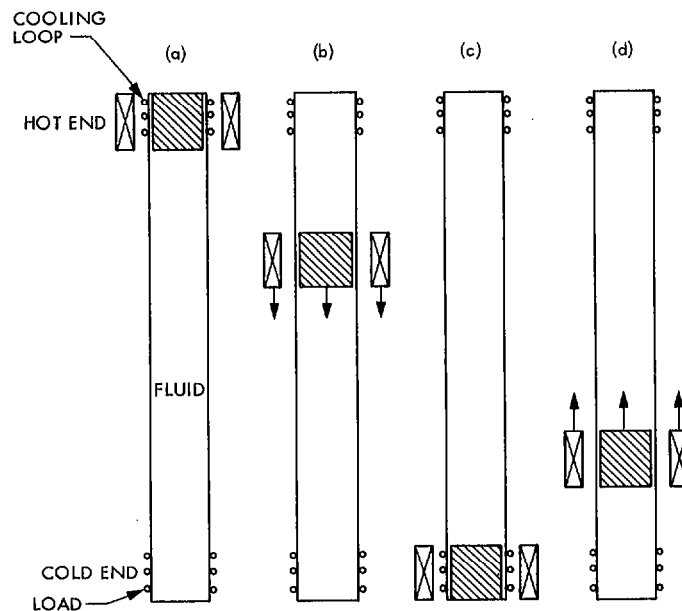


Fig. 4. Schematic diagram of magnetic Stirling cycle. The processes are (a) isothermal magnetization, (b) isofield cooling in regenerator, (c) isothermal demagnetization, and (d) zero (or low) field heating in regenerator (taken from Ref. 19)

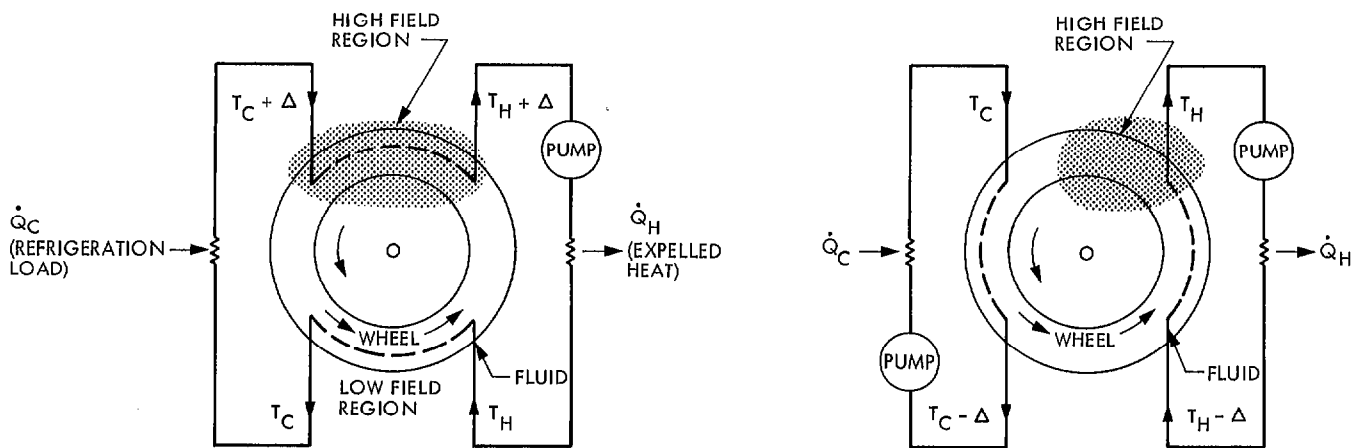


Fig. 5. Schematics of (a) Stirling-cycle wheel and (b) Carnot-cycle wheel refrigerators. The paramagnetic material forming the rim of the wheels rotates in a counterclockwise direction. The fluid flow is forced by a pump and its direction of motion is indicated by the arrows. The high magnetic field region is indicated by the shaded area. Radially across the wheel is the low field region. Δ represents the inherent temperature change of the working material upon entering and leaving the field (taken from Ref. 23)

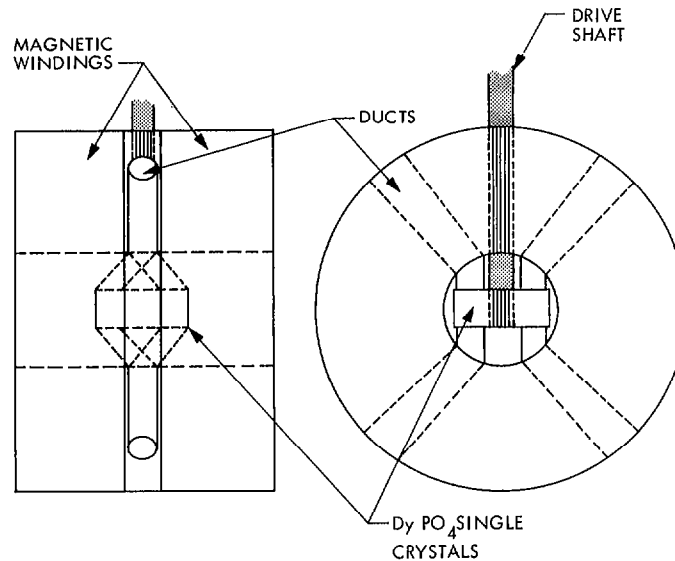


Fig. 6. Side and end views of a wheel of DyPO_4 single crystals inside a superconducting magnet. The drive shaft and ducts are also shown (taken from Ref. 11)